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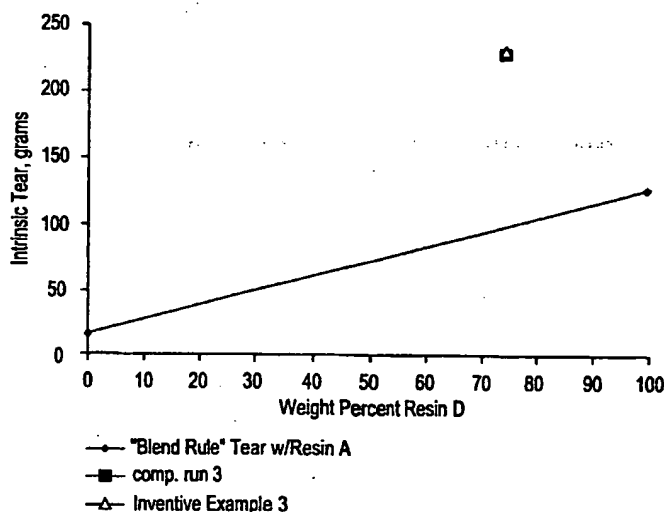
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[Continued on next page]

(54) Title: POLYMER BLEND AND FABRICATED ARTICLE MADE FROM DIVERSE ETHYLENE INTERPOLYMERS



(57) Abstract: This invention is directed to an ethylene polymer blend comprising at least two diverse ethylene interpolymers wherein one interpolymer has a lower number of carbons than the at least one other interpolymer. The ethylene polymer blend preferably comprises at least one homogeneously branched ethylene/ $\alpha$ -olefin interpolymer blended with at least one heterogeneously branched ethylene/ $\alpha$ -olefin interpolymer and is characterized as having a density greater than or equal to 0.90 g/cm<sup>3</sup> and in particularly preferred embodiments is further characterized as having an intrinsic tear value greater than or equal to 150 grams. The inventive ethylene polymer blend can be used to make various fabricated articles, especially extruded forms and most especially films such as high strength thin gauge packaging film, impact resistant shrink film and heat sealable packaging film.

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## POLYMER BLEND AND FABRICATED ARTICLE MADE FROM DIVERSE ETHYLENE INTERPOLYMERS

This invention relates to an ethylene polymer blend made from or  
5 comprising at least two diverse ethylene interpolymers wherein one  
interpolymer has a lower number of carbons than the at least one other  
interpolymer. The ethylene polymer blend preferably comprises at least one  
homogeneously branched ethylene/ $\alpha$ -olefin interpolymer blended with at  
least one heterogeneously branched ethylene/ $\alpha$ -olefin interpolymer and is  
10 characterized as having a density greater than or equal to 0.90 g/cm<sup>3</sup> and  
an intrinsic tear value greater than or equal to 150 grams-force.

The inventive ethylene polymer blend can be used in various  
extruded forms and is particularly useful for film applications (for example,  
high strength thin gauge packaging film, impact resistant shrink film and  
15 heat sealable packaging film).

Film products fabricated from linear low density polyethylene  
(LLDPE) or high density polyethylene (HDPE) are widely used for packaging  
applications such as merchandise bags, shrink films, grocery sacks, and  
industrial liners. For these applications, films with excellent toughness  
20 properties (that is, high tensile, impact or tear strength or combinations of  
these) are desired to facilitate down-gauging or prevent premature failures  
during handling, distribution and use.

Previous attempts were made to optimize film tensile strength and  
yield strength by blending various heterogeneous polymers together on a  
25 theoretical basis. While such blends exhibited a synergistic response of  
increased the film yield strength, the film impact strength typically followed  
the rule of blend, often resulting in a "destructive synergism" (that is, the film  
impact strength was actually lower than film made from one of the two  
components used to make the blend).

For example, it is known that while improved modulus linear polyethylene polymer blends can be produced by blending high density polyethylene with a very low density polyethylene (VLDPE), the impact strength of these polymer blends typically follow the rule of blend (or the  
5 "blend rule"). That is, the final composition exhibits properties that are comparable to and predictable from weight average calculations based on the component polymers. For example, see FIG. 5 herein.

It is also known from U.S. Patent No. 5,677,383, that dramatic synergism results when polymer compositions are made from a  
10 homogeneously branched ethylene interpolymer having a high slope of strain hardening coefficient melt-blended with a heterogeneously branched ethylene polymer. Although U.S. Patent No. 5,677,383 broadly discloses the homogeneously branched ethylene interpolymer and the heterogeneously branched ethylene polymer blend composition can  
15 comprise various comonomers, all of the presented examples consist of higher alpha olefin interpolymer combinations. Moreover, U.S. Patent No. 5,677,383 focuses on the slope of strain hardening coefficient of the component polymers and does not explicitly disclose the slope of strain hardening coefficient of the final blend combinations.

It is also known that, at the equivalent densities, interpolymers  
20 comprised of higher alpha olefins provide improved toughness properties as compared to lower alpha olefin interpolymers. For example, at a density of about 0.935 g/cc, an ethylene/1-octene copolymer will exhibit intrinsic tear that is about two times higher than that of an ethylene/1-butene copolymer  
25 having about the same density.

While it is well established that higher alpha olefin interpolymers provide superior toughness, but with periodic scarcities in the availability of higher alpha olefin comonomers, resin producers and fabricators desire broader polymer options for providing polymer compositions characterized  
30 by excellent toughness properties. The need is especially great in regard to

1-octene, which is a higher alpha olefin that frequently tends to be in short supply while the lower alpha olefin 1-butene generally tends to be abundantly available. For polymer blends comprising at least two higher alpha olefin interpolymers, there is a particular need to substitute at least one of the higher alpha olefin interpolymers with a lower alpha olefin interpolymers and still retain the excellent toughness properties characteristic of higher alpha olefin polymer blends. For polymer blends comprising at least two higher alpha olefin interpolymers where one is a homogeneously branched ethylene interpolymers and the other is a heterogeneously branched ethylene polymer, there is an especially particular need to substitute at least one of the higher alpha olefin interpolymers with a lower alpha olefin interpolymers and still retain the excellent toughness properties characteristic of these polymer blends.

Surprisingly, we have now discovered that within a certain narrow density range, tailored combinations of at least one interpolymers comprised of a lower carbon comonomer blended with at least one interpolymers comprised of a higher carbon comonomer can provide toughness properties comparable to polymer blends comprised of two interpolymers, each having the same higher carbon comonomer or, alternatively, comparable to single interpolymers compositions where the interpolymers comprises a higher carbon comonomer.

FIG.1A and 1B are diagrams of test dogbones used for determining the slope of strain hardening coefficient.

FIG. 2 shows the various regions of a typical tensile curve (as load (in pounds) versus extension (in inches)) and the particular region used to determine the slope of strain hardening.

FIG. 3 shows the relationship between intrinsic tear (in grams) and the slope of strain hardening coefficient (SHC) for two homogeneously branched ethylene/1-octene copolymers and one heterogeneously branched ethylene/1-octene copolymer.

FIG. 4 shows the relationship between intrinsic tear (in grams) and the overall density (in grams per cubic centimeter) for various single polymer and polymer blend compositions, including inventive interpolymer combinations.

5           FIG. 5 shows, in regard to intrinsic tear (in grams), how dissimilar and similar interpolymer combinations compare to a "blend rule" polymer blend of a homogeneously branched ethylene/1-butene copolymer (having a 0.8714 g/cc density) and heterogeneously branched ethylene/1-octene copolymer (having a 0.934 g/cc density).

10           FIG. 6 shows, in regard to intrinsic tear (in grams), how dissimilar and similar interpolymer combinations compare to a "blend rule" polymer blend of a homogeneously branched ethylene/1-octene copolymer (having a 0.8688 g/cc density) and heterogeneously branched ethylene/1-octene copolymer (having a 0.934 g/cc density).

15           FIG. 7 shows, in regard to intrinsic tear (in grams), how dissimilar and similar interpolymer combinations compare to a "blend rule" polymer blend of a homogeneously branched ethylene/1-octene copolymer (having a 0.909 g/cc density) and heterogeneously branched ethylene/1-butene copolymer (having a 0.936 g/cc density).

20           FIG. 8 shows, in regard to intrinsic tear (in grams), how dissimilar and similar interpolymer combinations compare to a "blend rule" polymer blend of a homogeneously branched ethylene/1-octene copolymer (having a 0.909 g/cc density) and heterogeneously branched ethylene/1-octene copolymer (having a 0.935 g/cc density).

25           FIG. 9 shows, in regard to intrinsic tear (in grams), how dissimilar and similar interpolymer combinations compare to a "blend rule" polymer blend of a homogeneously branched ethylene/1-octene copolymer (having a 0.8714 g/cc density) and heterogeneously branched ethylene/1-octene copolymer (having a 0.920 g/cc density).

Formulated ethylene interpolymer compositions comprised of dissimilar interpolymers where at least one interpolymer comprises a lower carbon comonomer relative to the other interpolymer (or interpolymers), have now been discovered to have improved physical and mechanical strength and are  
5 useful in making fabricated articles. These novel compositions exhibit surprisingly good toughness properties which are comparable to synergistic interpolymer compositions comprised of two interpolymers, each having the same higher carbon comonomer; or comparable to "blend rule" polymer blends comprised two interpolymers, each having the same higher carbon  
10 comonomer; or comparable to single polymer compositions comprised higher carbon comonomers.

The broad aspect of the invention is a diverse interpolymer composition comprising

(A) from 5 percent (by weight of the total composition) to 95 percent  
15 (by weight of the total composition) of at least one first ethylene interpolymer, and

(B) from 95 percent (by weight of the total composition) to 5 percent (by weight of the total composition) of at least one second ethylene interpolymer;

20 wherein the first interpolymer is characterized as comprising a different comonomer relative to the second interpolymer, one interpolymer is a homogeneously branched ethylene interpolymer and the other interpolymer is a heterogeneously branched ethylene interpolymer or a homogeneously branched ethylene interpolymer, and the composition is characterized as  
25 having a density greater than or equal to 0.90 g/cm<sup>3</sup>.

In another aspect, the diverse interpolymer composition comprises

(A) from 5 percent (by weight of the total composition) to 95 percent (by weight of the total composition) of at least one first ethylene interpolymer, and

(B) from 95 percent (by weight of the total composition) to 5 percent (by weight of the total composition) of at least one second ethylene interpolymer;

wherein the first interpolymer is characterized as comprising a different  
5 comonomer relative to the second interpolymer and as having a lower density relative to the second interpolymer; the second interpolymer is characterized as having a density less than or equal to 0.95 g/cm<sup>3</sup>;

wherein one interpolymer is a homogeneously branched ethylene interpolymer and the other interpolymer is a heterogeneously branched  
10 ethylene interpolymer or a homogeneously branched ethylene interpolymer; and

wherein the composition is characterized as having a density in the range of from greater than or equal to 0.90 g/cc to 0.935 g/cc.

In certain preferred embodiments of the present invention, preferably, at  
15 least the first interpolymer or the second interpolymer or both have a slope of strain hardening coefficient greater than or equal to 1.3, more preferably greater than or equal to 1.5. In other preferred embodiments, the diverse interpolymer composition has an intrinsic tear value greater than or equal to 150 grams, preferably greater than or equal to 200 grams, more preferably  
20 greater than or equal 225 grams, or a slope of strain hardening coefficient greater than or equal to 0.85, preferably greater than or equal to 0.95, more preferably greater than or equal to 1.

In still other preferred embodiments, especially when balanced properties are desired, the first interpolymer comprises a lower carbon  
25 comonomer. More preferably, the first interpolymer comprises a lower carbon comonomer and is a homogeneously branched ethylene interpolymer. Most preferably, the first interpolymer comprises a lower carbon comonomer and is a substantially linear ethylene interpolymer.

The term "lower carbon comonomer" is used herein in reference to a  
30 comonomer that has a lower total number of carbons relative to another



comonomer. Similarly, the term "higher carbon comonomer" is used herein in reference to a comonomer that has a higher total number of carbons relative to another comonomer. The term "higher carbon comonomer" is a generic reference that embraces the term "higher lefin" and the term "lower carbon comonomer" is a generic term that embraces the term "lower lefin". Thus, for example, 1-octene is would be a higher carbon comonomer (as well as a higher lefin) relative to 1-butene and, conversely, 1-butene would be a lower carbon comonomer (as well as a lower lefin) relative to 1-octene. Also, 1-octene would be a "different comonomer" (as this term is used herein) relative to 1-butene.

10 By the term "comparable toughness properties" it is meant that the intrinsic tear of the inventive diverse interpolymer composition, as determined in accordance with ASTM D-1922, Method A), is within 35 percent, preferably 25 percent, more preferably 12 percent of the intrinsic tear of a comparative "blend rule" polymer blend wherein the compared polymer blend is comprised two  
15 interpolymers, each having the same higher carbon comonomer. Comparable intrinsic tear performance is considered to be a surprisingly substantial improvement because ordinarily interpolymers comprised higher carbon comonomers are expected to exhibit vastly superior toughness properties over interpolymers based on lower carbon comonomer.

20 The term "blend rule" as discussed above is used herein to refer to polymer blend property results attainable from fractional weight average calculations based on the properties of the component polymers of the polymer blend.

The density of the inventive diverse interpolymer composition (as  
25 measured in accordance with ASTM D-792) is generally from greater than or equal to  $0.90 \text{ g/cm}^3$ . Preferably, in the range of from greater than or equal to  $0.90 \text{ g/cm}^3$  to  $0.935 \text{ g/cm}^3$ , more preferably from greater than or equal to  $0.90 \text{ g/cm}^3$  to less than or equal to  $0.925 \text{ g/cm}^3$ , most preferably from  $0.905 \text{ g/cm}^3$  to  $0.925 \text{ g/cm}^3$ , and especially from  $0.905 \text{ g/cm}^3$  to than  $0.92 \text{ g/cm}^3$ .

The  $I_2$  melt index of the inventive diverse interpolymer composition is preferably in the range of from 0.1 to 100 g/10 minutes, more preferably in the range of from 0.2 to 10 g/10 minutes, most preferably less than or equal to 2.5 g/10 minutes.

- 5        The inventive diverse interpolymer composition can be alternately or further characterized as having an intrinsic tear (as measured in accordance with ASTM D-1922, Method A) preferably greater than or equal to 150 grams, more preferably greater than or equal to 200 grams and most preferably greater than or equal to 225 grams, and especially greater than or equal to 250 grams.
- 10       Preferably, the carbon difference between the at least one comonomer of the first interpolymer and the at least one comonomer of the second interpolymer is greater than 2, especially greater than or equal to 3, most especially greater than or equal to 4. Especially preferred are copolymer combinations (that is, each interpolymer comprises ethylene with only one other
- 15       comonomer). More especially preferred are copolymer combinations comprising 1-octene (for example combinations of 1-octene with 1-pentene, 1-octene with 1-butene and 1-octene with propylene). Most especially preferred are copolymer combinations comprising 1-octene and 1-butene (for example, a substantially linear ethylene/1-octene copolymer blended with a
- 20       heterogeneously branched ethylene/1-butene copolymer).

The homogeneously branched ethylene interpolymer is preferably a substantially linear ethylene/ $\alpha$ -olefin or diolefin interpolymer, as described in US Patent No. 5,272,236 and US Patent No. 5,278,272. The homogeneously branched ethylene interpolymer can also be a linear ethylene interpolymer

25       (that is, characterized as having no long chain branching and certain less than 0.01 long chain branch per 1000 carbon atoms) as described, for example, in U.S. Patent No. 3,645,992 (Elston).

Substantially linear ethylene interpolymers are not "linear" polymers in the traditional sense of the term and as used to describe linear low density

30       polyethylene (for example, Ziegler polymerized linear low density polyethylene

(LLDPE)). Nor are substantially linear ethylene interpolymers highly branched polymers, as used to describe high pressure, free-radical initiated low density polyethylene (LDPE).

The homogeneously branched ethylene interpolymers useful for forming the diverse interpolymer composition described herein are those in which the comonomer is randomly distributed within a given interpolymer molecule and wherein substantially all of the interpolymer molecules have the same ethylene/comonomer ratio within that interpolymer. The homogeneity of homogeneously branched ethylene interpolymers (either substantially linear ethylene interpolymers or homogeneously branched linear ethylene interpolymers) is typically described by the SCBDI (Short Chain Branch Distribution Index) or CDBI (Composition Distribution Branch Index).

SCBDI is defined as the weight percent of the polymer molecules having a comonomer content within 50 percent of the median total molar comonomer content and represents a comparison of the comonomer distribution in the interpolymer to the comonomer distribution expected for a Bernoullian distribution. The SCBDI of an interpolymer can be readily calculated from TREF as described, for example, by Wild et al., Journal of Polymer Science, Poly. Phys. Ed., Vol. 20, p. 441 (1982), or in US Patent 4,798,081; 5,008,204; or by L. D. Cady, "The Role of Comonomer Type and Distribution in LLDPE Product Performance," SPE Regional Technical Conference, Quaker Square Hilton, Akron, Ohio, October 1-2, pp. 107-119 (1985).

The preferred TREF technique does not include purge quantities in SCBDI calculations. More preferably, the comonomer distribution of the interpolymer and SCBDI are determined using  $^{13}\text{C}$  NMR analysis in accordance with techniques described in US Patent No. 5,292,845; US Patent No. 4,798,081; U.S. Patent No. 5,089,321 and by J. C. Randall, Rev. Macromol. Chem. Phys., C29, pp. 201-317.

In analytical temperature rising elution fractionation analysis (as described in US Patent No. 4,798,081 and abbreviated herein as "ATREF"), the film or composition to be analyzed is dissolved in a suitable hot solvent (for example, trichlorobenzene) and allowed to crystallize in a column containing an inert support (stainless steel shot) by slowly reducing the temperature. The column is equipped with both a refractive index detector and a differential viscometer (DV) detector. An ATREF-DV chromatogram curve is then generated by eluting the crystallized polymer sample from the column by slowly increasing the temperature of the eluting solvent (trichlorobenzene). The ATREF curve is also frequently called the short chain branching distribution (SCBD), since it indicates how evenly the comonomer (for example, octene) is distributed throughout the sample in that as elution temperature decreases, comonomer content increases. The refractive index detector provides the short chain distribution information and the differential viscometer detector provides an estimate of the viscosity average molecular weight. The short chain branching distribution and other compositional information can also be determined using crystallization analysis fractionation such as the CRYSTAF fractional analysis package available commercially from PolymerChar, Valencia, Spain.

The SCBDI for the homogeneously branched linear ethylene interpolymer and the substantially linear ethylene interpolymers used in the present invention is preferably greater than 50 percent, especially greater than 70 percent and most especially greater than 90 percent. That is, by the term "homogeneously branched ethylene interpolymer" it is meant an ethylene interpolymer having a SCBDI greater than 50 percent.

Preferred homogeneously branched ethylene interpolymers useful in this invention also essentially lack a measurable "linear" polymer fraction (also referred to as a "high density" or "crystalline" polymer fraction) as measured by the TREF technique (that is, preferred homogeneously branched ethylene interpolymers do not contain a polymer fraction with a degree of branching less

than or equal to 2 methyls/1000 carbons). More preferably, the homogeneously branched ethylene interpolymers also do not contain any highly short chain branched fraction (that is, more preferred homogeneously branched ethylene interpolymers do not contain a polymer fraction with a  
5 degree of branching equal to or more than 30 methyls/1000 carbons). Although suitable for use in the present invention and are not preferred (or more preferred), commercial examples of homogeneously branched ethylene interpolymers having a "linear" polymer fraction include certain grades of EXCEED resins supplied by Exxon Chemical Company. Commercial  
10 examples of suitable homogeneously branched ethylene polymers believed not to possess a "linear" polymer fraction are EXACT resins supplied by Exxon chemical Company and TAFMER resins supplied by Mitsui Chemical Company.

The term "interpolymer" is used herein to indicate, for example, a  
15 copolymer or a terpolymer. That is, at least one other comonomer is polymerized with ethylene to make the interpolymer.

The homogeneously branched ethylene interpolymers and heterogeneously branched ethylene interpolymers for use in the present invention typically are interpolymers of ethylene with at least one C<sub>3</sub>-  
20 C<sub>20</sub>  $\alpha$ -olefin or C<sub>4</sub>-C<sub>18</sub> diolefin. Copolymers of ethylene and 1-octene are especially preferred.

Ethylene copolymerized with two or more comonomers can also be used to make the homogeneously branched substantially linear interpolymers useful in this invention. Preferred comonomers include the C<sub>3</sub>-C<sub>20</sub>  $\alpha$ -olefins,  
25 especially propylene, isobutylene, 1-butene, 1-hexene, 4-methyl-1-pentene, 1-heptene, 1-octene, 1-nonene, and 1-decene, more preferably 1-butene, 1-hexene, heptene and 1-octene.

The term "linear ethylene interpolymer" means that the interpolymer does not have long chain branching. That is, the linear ethylene interpolymer  
30 has an absence of long chain branching as described above. The term "linear

ethylene interpolymer" does not refer to high pressure branched, free-radical initiated polyethylene (LDPE) which is known to those skilled in the art to have numerous long chain branches. However, the comonomer branching distribution (i.e. short chain branching distribution) of homogeneously branched linear ethylene interpolymers is the same or substantially the same as that described for the substantially linear ethylene/ interpolymers.

Substantially linear ethylene interpolymers have a single melting point, as opposed to traditional heterogeneously branched Ziegler polymerized ethylene interpolymers which have two or more melting points, as determined using differential scanning calorimetry (DSC). Most (but not all) homogeneously branched linear ethylene interpolymers also have a single DSC melting point. Those homogeneously branched linear ethylene interpolymers that have a "linear" polymer fraction in TREF testing, generally have more than one DSC melting point. Homogeneously branched ethylene polymers having a single DSC melting point in the temperature range of  $-30^{\circ}$  to  $150^{\circ}\text{C}$  are preferred for use in the present invention.

Preferably, the first interpolymer has a lower density than the second interpolymer. More preferably, the density of the first interpolymer, as measured in accordance with ASTM D-792, for use in the present invention is especially from  $0.89\text{ g/cm}^3$  to  $0.935\text{ g/cm}^3$ , more especially from  $0.90\text{ g/cm}^3$  to  $0.925\text{ g/cm}^3$ , most especially from  $0.905\text{ g/cm}^3$  to  $0.925\text{ g/cm}^3$ , and most particularly from  $0.905\text{ g/cm}^3$  to than  $0.92\text{ g/cm}^3$ .

The amount of the first interpolymer and second interpolymer incorporated into the inventive diverse interpolymer composition varies depending upon the desired melt index and targeted density of the final composition. But preferably, the interpolymer composition comprises from greater than 20 weight percent to 95 weight percent (based on the total weight of the composition) of the first interpolymer and from less than 80 weight percent to 5 weight percent (based on the total weight of the composition) of the second interpolymer. More preferably, the interpolymer composition

comprises from greater than 25 weight percent to 85 weight percent (based on the total weight of the composition) of the first interpolymer and from less than 75 weight percent to 15 weight percent (based on the total weight of the composition) of the second interpolymer.

- 5           The molecular weight of the homogeneously branched ethylene interpolymer or the heterogeneously branched ethylene interpolymer for use in the present invention is conveniently indicated using a melt index measurement according to ASTM D-1238, Condition 190°C/2.16 kg (formerly known as "Condition (E)" and also known as  $I_2$ ). Melt index is inversely proportional to
- 10           the molecular weight of the polymer. Thus, the higher the molecular weight, the lower the melt index, although the relationship is not linear. The lower melt index limit for the ethylene interpolymers useful herein is preferably about 0.001 grams/10 minutes (g/10 min). The upper melt index limit is preferably 10 g/10 min, preferably less than 1 g/10 min, and especially less than 0.5 g/10 min.
- 15           Most preferably, the first interpolymer is a homogeneously branched ethylene interpolymer having an  $I_2$  melt index equal to or lower than that of the second interpolymer.

Another measurement useful in characterizing the molecular weight of the ethylene interpolymers is conveniently indicated using a melt index

20           measurement according to ASTM D-1238, Condition 190°C/10 kg (formerly known as "Condition (N)" and also known as  $I_{10}$ ). The ratio of the  $I_{10}$  and  $I_2$  melt index terms is the melt flow ratio and is designated as  $I_{10}/I_2$ .

Generally, the  $I_{10}/I_2$  ratio for the homogeneously branched linear ethylene interpolymers is about 5.6.

- 25           For the substantially linear ethylene interpolymers used in the inventive composition, the  $I_{10}/I_2$  ratio indicates the degree of long chain branching; that is, the higher the  $I_{10}/I_2$  ratio, the more long chain branching in the interpolymer. Generally, the  $I_{10}/I_2$  ratio of the branched substantially linear ethylene interpolymers is at least about 6, preferably at least about 7, especially at least

about 8 or above. For the homogeneously branched substantially linear ethylene interpolymers, the higher the  $I_{10}/I_2$  ratio, the better the processability.

The rheological processing index (PI), onset of surface and gross melt fracture are measured using a gas extrusion rheometer (GER). The GER is described by M. Shida, R.N. Shroff and L.V. Cancio in Polym. Eng. Sci., Vol. 17, no. 11, p. 770 (1977), and in Rheometers for Molten Plastics by John Dealy, published by Van Nostrand Reinhold Co. (1982) on pages 97-99. PI testing is performed at a temperature of 190°C, at nitrogen pressure of 2500 psig using 0.0296 inch diameter, 20:1 L/D die having an entrance angle of 180°. The GER processing index is calculated in millipoise units from the following equation:

$$PI = 2.15 \times 10^6 \text{ dynes/cm}^2 / (1000 \times \text{shear rate}),$$

where:  $2.15 \times 10^6$  dynes/cm<sup>2</sup> is the shear stress at 2500 psi, and the shear rate is the shear rate at the wall as represented by the following equation:

$$32 Q' / (60 \text{ sec/min})(0.745)(\text{Diameter} \times 2.54 \text{ cm/in})^3, \text{ where:}$$

$Q'$  is the extrusion rate (gms/min),

0.745 is the melt density of polyethylene (gm/cm<sup>3</sup>), and

Diameter is the orifice diameter of the capillary (inches).

The PI is the apparent viscosity of a material measured at apparent shear stress of  $2.15 \times 10^6$  dyne/cm<sup>2</sup>.

For the substantially linear ethylene interpolymers used herein, the PI is preferably less than or equal to 70 percent of that of a comparative linear ethylene interpolymers; that is, a homogeneously branched linear ethylene interpolymers having about the same  $I_2$  and  $M_w/M_n$ .

An apparent shear stress vs. apparent shear rate plot is used to identify the melt fracture phenomena. According to Ramamurthy in Journal of Rheology, 30(2), pages 337-357 (1986), above a certain critical flow rate, the observed extrudate irregularities may be broadly classified into two main types: surface melt fracture and gross melt fracture.



Surface melt fracture occurs under apparently steady flow conditions and ranges in detail from loss of specular gloss to the more severe form of "sharkskin". In this disclosure, the onset of surface melt fracture is characterized at the beginning of losing extrudate gloss at which the surface roughness of extrudate can only be detected by 40x magnification. The critical shear rate at onset of surface melt fracture for the substantially linear olefin polymers is at least 50 percent greater than the critical shear rate at the onset of surface melt fracture of a comparative linear ethylene interpolymer; that is, a homogeneously branched linear ethylene interpolymer having about the same  $I_2$  and  $M_w/M_n$ .

Gross melt fracture occurs at unsteady flow conditions and ranges in detail from regular (alternating rough and smooth, helical, etc.) to random distortions. For commercial acceptability, (for example, in blown film products), surface defects should be minimal, if not absent. The critical shear rate at onset of surface melt fracture (OSMF) and onset of gross melt fracture (OGMF) will be used herein based on the changes of surface roughness and configurations of the extrudates extruded by a GER.

GER testing for melt fracture determinations is performed at a temperature of 190°C, at nitrogen pressures between 5250 to 500 psig using a 0.0296 inch diameter, 20:1 L/D die having an entrance angle of 180°.

Substantially linear ethylene interpolymers suitable for use herein are characterized as having:

- (a) a melt flow ratio,  $I_{10}/I_2$  5.63,
- (b) a molecular weight distribution,  $M_w/M_n$ , as determined by gel permeation chromatography and defined by the equation:  
$$(M_w/M_n) \leq (I_{10}/I_2) - 4.63,$$
- (c) a gas extrusion rheology such that the critical shear rate at onset of surface melt fracture for the substantially linear ethylene interpolymer is at least 50 percent greater than the critical shear rate at the onset of surface melt fracture for a comparative

5

homogeneously branched linear ethylene interpolmer, wherein the substantially linear ethylene interpolmer and the homogeneously branched linear ethylene interpolmer comprise the same comonomer or comonomers, the homogeneously branched linear ethylene interpolmer has an  $I_2$  and  $M_w/M_n$

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within ten percent of the substantially linear ethylene interpolmer and wherein the respective critical shear rates of the interpolmers are measured at the same melt temperature, under the same nitrogen load using a gas extrusion rheometer, and (d) a single differential scanning calorimetry, DSC, melting peak between

-30° and 150°C.

15

The molecular weight distribution of the ethylene interpolmers is analyzed by gel permeation chromatography (GPC) on a Waters 150C high temperature chromatographic unit equipped with three mixed porosity columns (Polymer Laboratories  $10^3$ ,  $10^4$ ,  $10^5$ , and  $10^6$ ), operating at a system temperature of 140°C. The solvent is 1,2,4-trichlorobenzene, from which 0.3 percent by weight solutions of the samples are prepared for injection. The flow rate is 1.0 milliliter/minute and the injection size is 200 microliters. A differential refractometer is being used as the detector.

20

The molecular weight determination with respect to the polymer backbone is deduced by using narrow molecular weight distribution polystyrene standards (from Polymer Laboratories) in conjunction with their elution volumes. The equivalent polyethylene molecular weights are determined by using appropriate Mark-Houwink coefficients for polyethylene and polystyrene (as described by Williams and Ward in Journal of Polymer Science, Polymer Letters, Vol. 6, p. 621, 1968) to derive the following equation:

25

$$M_{\text{polyethylene}} = a * (M_{\text{polystyrene}})^b.$$

30

In this equation,  $a = 0.4316$  and  $b = 1.0$ . Weight average molecular weight,  $M_w$ , is calculated in the usual manner according to the following formula:  $M_j = (\sum w_i(M_i)^j)^{1/j}$ ; where  $w_i$  is the weight fraction of the molecules with molecular weight  $M_i$  eluting from the GPC column in fraction  $i$  and  $j = 1$  when calculating  $M_w$  and  $j = -1$  when calculating  $M_n$ .

For the homogeneously branched ethylene interpolymers used herein, the molecular weight distribution ( $M_w/M_n$ ) is thought not to be critical but preferably the molecular weight distribution, as determined using gel permeation chromatography (GPC), is from 1.8 to 2.8, more preferably from 1.89 to 2.2 and especially less than 2.

The slope of strain hardening is measured by compression molding a plaque from the polymer to be tested. Typically, the plaque is molded at about 190°C for 4 minutes under almost no pressure and then pressed for 3 minutes under a pressure of about 20,000 psi (138 MPa). The plaque is then allowed to cool at about 8°C/minute under about 2,000 psi pressure (13.8 MPa). The molded plaque has a thickness of from 0.005 to 0.007 inches (0.013-0.018 cm). The plaque is then cut into a dogbone shaped test piece using a suitable die. A suitable test piece is 0.315 inches (0.8 cm) wide and 1.063 inches (2.7 cm) long. The start of the curved portion of the dogbone shape begins at 0.315 inches (0.8 cm) from each end of the sample and gently curves (that is, tapers) to an interior width of from 0.079 to 0.090 inches (0.2-0.23 cm). The curve ends at a point 0.118 inches (0.3 cm) from the start of the curve such that the interior portion of the dogbone test piece has a length of 0.197 inches (0.5 cm). FIG. 1A and 1B show scale diagrams of suitable test dogbones.

The tensile properties of the test sample are tested on an Instron Tensile Tester at a crosshead speed of 1 inch/minute (2.5 cm/min.). The slope of strain hardening is calculated from the resulting tensile curve by drawing a line parallel to the strain hardening region (that is a 10 percent secant tangent line). The strain hardening region occurs after the sample has pulled its initial load (usually with little or no elongation during the initial load) and after the sample

has gone through a slight drawing stage (usually with little or no increase in load, but with increasing extension (that is, strain)). In the strain hardening region, the load and the extension of the sample both continue to increase up to the failure point.

- 5           FIG. 2 shows a tensile curve with the various stages of strain hardening as well as the slope of strain hardening. The slope of strain hardening is conveniently taken as the line representing a 10 percent secant tangent which is calculated from the failure point to the point at 10 percent extension before the failure point (where 10 percent extension before is equal to 90 percent of  
10   the total extension or strain).

A more precise methodology for calculating the slope of strain hardening is performing linear regression analysis using the tensile curve datapoints that represent the last 10 percent extension before the failure point.

- The slope of strain hardening coefficient (SHC) is calculated according to  
15   the equation:  $SHC = (\text{slope of strain hardening}) * (I_2)^{0.25}$

          where  $I_2$  = melt index in grams/10 minutes, as determined in accordance with ASTM D-1238, Condition 190°C/2.16 kg.

- For the first interpolymer or the second interpolymer or both, preferably the SHC is greater than or equal to 1.3, more preferably greater than or equal  
20   to 1.5, especially in the range of from 1.3 to 10, and most especially in the range of from 1.5 to 5. More preferably, the inventive diverse interpolymer composition itself is characterized as having a SHC greater than or equal to  
0.85, most preferably greater than or equal to 0.95, most especially greater than or equal to 1.

- 25           The second ethylene interpolymer can be a homogeneously branched ethylene interpolymer or a heterogeneously branched ethylene interpolymer. But, preferably, the second interpolymer is a heterogeneously branched (for example, Ziegler polymerized) interpolymer of ethylene with at least one  $C_3$ - $C_{20}$   $\alpha$ -olefin (for example, linear low density polyethylene (LLDPE)). By the  
30   term "heterogeneously branched ethylene interpolymer" it is meant the

ethylene interpolymer has a SCBDI of less than 50 percent, more preferably less than 30 percent.

Heterogeneously branched ethylene interpolymers differ from the homogeneously branched ethylene interpolymers primarily in their branching distribution. For example, heterogeneously branched LLDPE polymers have a distribution of branching, including a highly branched portion (similar to a very low density polyethylene), a medium branched portion (similar to a medium branched polyethylene) and an essentially linear portion (similar to linear homopolymer polyethylene). The amount of each of these fractions varies depending upon the whole polymer properties desired. For example, linear homopolymer polyethylene has neither branched nor highly branched fractions, but is linear. A very low density heterogeneous polyethylene having a density from  $0.9 \text{ g/cm}^3$  to  $0.915 \text{ g/cm}^3$  (such as ATTANE<sup>TM</sup> copolymers, sold by The Dow Chemical Company and FLEXOMER<sup>®</sup> sold by Union Carbide Corporation) has a higher percentage of the highly short chain branched fraction, thus lowering the density of the whole polymer.

Heterogeneously branched ethylene interpolymers (such as DOWLEX<sup>®</sup> LLDPE resins sold by The Dow Chemical Company) have lower amounts of the highly branched fraction, but has a greater amount of the medium branched fraction. While ATREF will show homogeneously branched ethylene interpolymers typically are characterized by a single relatively narrow peak at some elution temperature, heterogeneously branched ethylene interpolymers like DOWLEX<sup>TM</sup> LLDPE 2045 resin have a broad branching distribution, as represented by the breadth of elution temperatures over which the polymer fractions elute. DOWLEX<sup>TM</sup> LLDPE 2045 resin also has a distinct peak at an elution temperature of about  $98^\circ\text{C}$ , indicating the "linear" polymer fraction of the whole polymer.

Preferably, the heterogeneously branched ethylene interpolymer is a copolymer of ethylene with a  $\text{C}_3\text{-C}_{20}$   $\alpha$ -olefin, wherein, more preferably, the copolymer has a density from  $0.93 \text{ g/cm}^3$  to  $0.95 \text{ g/cm}^3$ , and most preferably a

melt index ( $I_2$ ) from 0.1 g/10 min to 500 g/10 min. Most especially, the heterogeneously branched interpolymer will have a density greater than 0.920 g/cc and especially greater than or equal to 0.935 g/cm<sup>3</sup>.

In particularly preferred embodiments, the heterogeneously branched interpolymer will have a density of no more than 0.95 g/cm<sup>3</sup>, more preferably no more than 0.945 g/cm<sup>3</sup>, and most preferably no more than 0.94 g/cm<sup>3</sup>. When the density of the heterogeneously branched interpolymer is no more than about 0.95 g/cm<sup>3</sup>, the interpolymer will typically be characterized by a molecular weight distribution ( $M_w/M_n$ ) greater than 3, preferably at least 3.2, and more preferably at least 3.3. Further, when the density of the heterogeneously branched interpolymer is no more than about 0.95 g/cm<sup>3</sup>, the SCBDI, as defined above, is less than 50 percent and most commonly less than or equal to 45 percent.

The heterogeneously branched ethylene interpolymers used herein also have at least two melting points, as determined using Differential Scanning Calorimetry (DSC) between the temperature range of -30° to 150°C..

The compositions disclosed herein can be formed by any convenient method, including dry blending the individual components and subsequently melt blend or by pre-melt blend in a separate extruder (for example, a Banbury mixer, a Haake mixer, a Brabender internal mixer, or a twin screw extruder).

Another technique for making the compositions in-situ is disclosed in pending USSN 08/010,958, entitled Ethylene Interpolymerizations, which was filed January 29, 1993 in the names of Brian W.S. Kolthammer and Robert S. Cardwell. USSN 08/010,958 describes, *inter alia*, interpolymerizations of ethylene and C<sub>3</sub>-C<sub>20</sub>  $\alpha$ -olefins using a homogeneous catalyst in at least one reactor and a heterogeneous catalyst in at least one other reactor. The reactors can be operated sequentially or in parallel.

The compositions can also be made by fractionating a heterogeneous ethylene interpolymer into specific polymer fractions with each fraction having a

narrow composition (that is, branching) distribution, selecting the fraction having the specified properties (for example,  $\text{SHC} \geq 1.3$ ), and blending the selected fraction in the appropriate amounts with another ethylene polymer. This method is obviously not as economical as the *in-situ* interpolymerizations of USSN 08/010,958, but nonetheless can be used to obtain the present diverse interpolymer composition.

Many useful fabricated articles benefit from the diverse interpolymer composition disclosed herein. For example, molding operations can be used to form useful fabricated articles or parts from the compositions disclosed herein, including various injection molding processes (for example, that described in Modern Plastics Encyclopedia/89, Mid October 1988 Issue, Volume 65, Number 11, pp. 264-268, "Introduction to Injection Molding" by H. Randall Parker and on pp. 270-271, "Injection Molding Thermoplastics" by Michael W. Green) and blow molding processes (for example, that described in Modern Plastics Encyclopedia/89, Mid October 1988 Issue, Volume 65, Number 11, pp. 217-218, "Extrusion-Blow Molding" by Christopher Irwin), profile extrusion, calendering, pultrusion (for example, pipes) and combinations thereof. Rotomolded articles can also benefit from the novel compositions described herein. Rotomolding techniques are well known to those skilled in the art and include, for example, those described in Modern Plastics Encyclopedia/89, Mid October 1988 Issue, Volume 65, Number 11, pp. 296-301, "Rotational Molding" by R.L. Fair).

Fibers (for example, staple fibers, melt blown fibers or spunbonded fibers (using, for example, systems as disclosed in U.S. Patent No. 4,340,563; U.S. Patent No 4,663,220; U.S. Patent No 4,668,566; or U.S. Patent No 4,322,027), and gel spun fibers (for example, the system disclosed in U.S. Patent No 4,413,110)), both woven and nonwoven fabrics (for example, spunlaced fabrics disclosed in U.S. Patent No 3,485,706) or structures made from such fibers (including, for example, blends of these fibers with other fibers, for example, PET or cotton)) can also be made from the inventive diverse composition

disclosed herein. Elastic fibers comprising the inventive diverse interpolymer composition is a particularly preferred use.

Coatings, films and film structures (especially biaxial shrink films, lamination films and other films and film applications that require excellent toughness properties) particularly benefit from the inventive interpolymers compositions described herein. Such coatings, films and film structures can be made using conventional hot blown film fabrication techniques, cast extrusion, extrusion coating, coextrusion techniques, and biaxial orientation processes such as tenter frames or double bubble processes.

10. Conventional hot blown film processes are described, for example, in The Encyclopedia of Chemical Technology, Kirk-Othmer, Third Edition, John Wiley & Sons, New York, 1981, Vol. 16, pp. 416-417 and Vol. 18, pp. 191-192. Biaxial orientation film manufacturing processes (for example, a "double bubble" process or a "trapped bubble" process) useful fabricate shrink film from the inventive composition are described in U.S. Patent 3,456,044 (Pahlke), U.S. Patent No. 4,352,849 (Mueller), U.S. Patent No. 4,597,920 (Golike), U.S. Patent No. 4,820,557 (Warren), U.S. Patent No. 4,837,084 (Warren), U.S. Patent No. 4,865,902 (Golike et al.), U.S. Patent No. 4,927,708 (Herran et al.), U.S. Patent No. 4,952,451 (Mueller), U.S. Patent No. 4,963,419 (Lustig et al.), and U.S. Patent No. 5,059,481 (Lustig et al.) can also be used to make film structures from the inventive diverse interpolymer composition described herein. The film structures can also be made using the tenter-frame technique, such as that conventionally used for oriented polypropylene.

- Other multi-layer film manufacturing techniques for food packaging applications for which the present inventive composition is well suited are described in Packaging Foods With Plastics, by Wilmer A. Jenkins and James P. Harrington (1991), pp. 19-27, and in "Coextrusion Basics" by Thomas I. Butler, Film Extrusion Manual: Process, Materials, Properties pp. 31-80 (published by TAPPI Press (1992)). The films may be monolayer or multilayer films. The film made from the inventive interpolymer compositions can also be



coextruded with the other layer(s) or the film can be laminated onto another layer(s) in a secondary operation, such as that described in Packaging Foods With Plastics, by Wilmer A. Jenkins and James P. Harrington (1991) or that described in "Coextrusion For Barrier Packaging" by W.J. Schrenk and C.R.

- 5 Finch, Society of Plastics Engineers RETEC Proceedings, June 15-17 (1981), pp. 211-229. If a monolayer film is produced via tubular film (that is, blown film techniques) or flat die (that is, cast film) as described by K.R. Osborn and W.A. Jenkins in "Plastic Films, Technology and Packaging Applications" (Technomic Publishing Co., Inc. (1992)), then the film must go through an additional post-
- 10 extrusion step of adhesive or extrusion lamination to other packaging material layers to form a multilayer structure. If the film is a coextrusion of two or more layers (also described by Osborn and Jenkins), the film may still be laminated to additional layers of packaging materials, depending on the other physical requirements of the final film. "Laminations Vs. Coextrusion" by D. Dumbleton,
- 15 Converting Magazine (September 1992) also discusses lamination versus coextrusion. Monolayer and coextruded films can also go through other post extrusion techniques, such as a biaxial orientation process.

- Other film applications that can use the inventive diverse interpolymer composition include pouches (especially via vertical form-fill-seal equipment),
- 20 bags, sacks (for example, heavy-duty shipping sacks), stretch film (especially, stretch cling films), liners, diaper backsheets, tarps and canvases, bubble wrap, skin packaging films, blister packaging films and laminated films (for example, TYVEK film and decorative film).

- Extrusion coating is yet another technique for producing multilayer
- 25 structures using the inventive diverse interpolymer composition described herein. The inventive composition can comprise at least one layer of the structure. Similar to cast film, extrusion coating is a flat die technique. For example, a sealant layer (which the inventive interpolymer composition is also useful for) can be extrusion coated onto a substrate either in the form of a
- 30 monolayer or a coextruded extrudate.

Generally, for a multilayer film or packaging structure, the inventive diverse interpolymer composition described herein comprises at least one layer of the total multilayer structure. Other layers of the multilayer structure include but are not limited to barrier layers, and/or tie layers, and/or structural layers.

- 5 Various materials can be used for these layers, with some of them being used as more than one layer in the same film structure. Some of these materials include: foil, nylon, ethylene/vinyl alcohol (EVOH) copolymers, polyvinylidene chloride (PVdC), polyethylene terephthalate (PET), polypropylene (including oriented polypropylene (OPP)), ethylene/vinyl acetate (EVA) copolymers, 10 ethylene/acrylic acid (EAA) copolymers, ethylene/methacrylic acid (EMAA) copolymers, LLDPE, HDPE, LDPE, nylon, graft adhesive polymers (for example, maleic anhydride grafted polyethylene), and paper. Generally, the multilayer film structures comprise from 2 to about 7 layers.

- In one particularly preferred embodiment, the inventive composition is 15 used in a coextruded multilayer film wherein the film is made from and/or comprises a core or hidden layer made from or comprising a high pressure polyethylene resin especially LDPE) and skin or outer layers made from and/or comprising the inventive composition. Such coextruded multilayer films exhibit significantly improved comparative optical properties.

- 20 Additives such as antioxidants (for example, hindered phenolics (for example, Irganox® 1010 made by Ciba Geigy Corp.), phosphites (for example, Irgafos® 168 also made by Ciba Geigy Corp.)), cling additives (for example, PIB), antiblock additives, slip agents, pigments, fillers, or combinations thereof, can be included in the formulations, to the extent that they do not interfere with 25 the enhanced formulation properties discovered by Applicants.

Example 1

In an evaluation to provide polymer blends with excellent toughness properties, several different ethylene interpolymers were obtained. These interpolymers all had relatively low  $I_2$  melt indexes but they varied as to

5 homogeneity, comonomer type, and density. Table 1 lists the various ethylene polymers used in the evaluation.

Table 1

Resin	Interpolymer	$I_2$	Density,	Comonomer
	Type	Melt Index	g/cc	r Type
A	389-S homogeneously branched	0.82	0.871	1-butene
B	389-F homogeneously branched	0.97	0.869	1-octene
C	DOWLEX 2045 heterogeneously branched	1.0	0.920	1-octene
D	DOWLEX 2037 heterogeneously branched	2.4	0.934	1-octene
E	AFFINITY PL1840 homogeneously branched	0.97	0.909	1-octene
F	DOWLEX 2038 heterogeneously branched	0.93	0.935	1-octene
G	SCLAIR 14B heterogeneously branched	1.66	0.936	1-butene

10 The ethylene interpolymers were tumble dry blended at various weight percents and extruded at about 200°C on a 15 mm co-rotating twin-screw extruder to provide several two component polymer blends. The individual interpolymers and the interpolymer polymer blends were compression molded and tested to determine their respective intrinsic tear

15 performance. Also, Resin B, Resin E and Resin F were tested using

dogbones according to FIG. 1A to determine their respective slope of strain hardening coefficient. Table 2 provides the designation of the single polymer compositions, the description of the polymer blends and resultant properties for both.

5           From the data in Table 2, FIG. 3-9 were generated. FIG. 3 shows the relationship between intrinsic tear (in grams) and the slope of strain hardening coefficient (SHC) for two heterogeneously branched ethylene/1-octene copolymers (Resin C and Resin F) and one homogeneously branched ethylene/1-octene copolymer (Resin E). While this figure was  
10       generated using single or component interpolymers (and without being held to any particular theory), we believe FIG. 3 is also applicable to polymer blends, especially those polymer blends that are not characterized by synergistic toughness properties.

FIG. 4 shows the relationship between intrinsic tear (in grams) and  
15       the overall density (in grams per cubic centimeter) for various single component compositions and polymer blend compositions, including inventive interpolymers combinations (Inventive Example 1, Inventive Example 2 and Inventive Example 3). FIG. 4 shows Inventive Example 1  
(which had an intrinsic tear of 229.4 grams) exhibited toughness that  
20       approximated the "blend rule" response. That is, the intrinsic tear of Inventive Example 1 was within 29 percent of the value calculable from corresponding component interpolymers.

Similarly, FIG. 4 shows Inventive Example 2 (which had an intrinsic tear of 264.1 grams) exhibited toughness that approximated the "blend  
25       rule". The intrinsic tear of Inventive Example 2 was within 33 percent of that predictable based on fractional weight average calculations using corresponding interpolymers. Inventive Example 2 was also within 33 percent of the intrinsic tear performance of a polymer blend made of two interpolymers, each comprised of the same comonomer (comparative run  
30       2).

Table 2

Example	Composition	Overall Density, g/cm <sup>3</sup>	Intrinsic Tear, gm-force	SHC	Yield Stress, psi	Yield Stress, STD, psi	Ultimate Stress, psi	Ultimate Stress, STD, psi	Energy at Break, psi	Energy at Break, STD, psi
Inv. Ex 1	25% Resin E + 75% Resin G	0.9303	154.5	ND	2,289	52	3,288	255	341	27
Comp Run 1*	25% Resin E + 75% Resin F	0.9288	271.2	ND	2,284	33	4,485	200	407	30
Inv. Ex 2	50% Resin E + 50% Resin G	0.9233	264.1	ND	1,824	37	3,999	145	385	19
Comp Run 2*	50% Resin E + 50% Resin F	0.9230	350.4	ND	1,770	30	3,958	426	328	46
Inv. Ex 3	25% Resin A + 75% Resin D	0.9206	229.4	ND	1,752	15	3,541	229	346	27
Comp Run 3*	25% Resin B + 75% Resin D	0.9181	227.0	ND	1,702	52	3,481	172	325	26
Comp Run 4*	45% Resin A + 55% Resin C	0.8992	172.6	ND	683	32	2,774	274	215	29
Comp Run 5*	45% Resin B + 55% Resin C	0.8964	281.2	ND	708	24	2,623	156	217	16
Comp Run 6*	100% Resin A	0.8714	14.8	ND	139	3	1,463	99	121	11
Comp Run 7*	100% Resin B	0.8688	35.9	ND	131	7	1,479	78	105	6
Comp Run 8*	100% Resin C	0.920	405.6	1.5	1,637	38	3,813	116	307	13
Comp Run 9*	100% Resin D	0.934	125.7	ND	2,735	69	4,043	167	382	20
Comp Run 10	100% Resin E	0.909	410.7	2.1	1,030	21	3,882	208	289	23
Comp Run 11*	100% Resin F	0.935	124.9	0.55	2,781	49	4,335	558	406	67
Comp Run 12	100% Resin G	0.936	60.0	ND	2,566	63	2,327	80	200	11

\*not an example of the present invention; provided for comparison only. ND denotes "not determined". STD denotes standard deviation.

FIG. 4 also shows Inventive Example 3 (which had an intrinsic tear of 229.4 grams) exhibited toughness that was equivalent to that of a polymer blend made of two interpolymers, each comprised of the same comonomer (comparative run 3).

5           The results for Inventive Example 1, Inventive Example 2 and Inventive Example 3 were a surprising unexpected result. That is these two-component interpolymer blends all comprised or were made from one ethylene interpolymer which was comprised of or made from a lower  $\alpha$ -olefin while the "blend rule" comparison in FIG. 4 was generated from a  
10 higher  $\alpha$ -olefin single polymer and polymer blends that consisted of two higher  $\alpha$ -olefin interpolymers and higher  $\alpha$ -olefin interpolymers are well known to exhibit improved toughness relative to lower  $\alpha$ -olefin interpolymers. In short, the fact that comparable or equivalent toughness can be accomplished using combinations with a lower  $\alpha$ -olefin is extremely  
15 surprising and unexpected.

FIG. 5 and FIG. 6 show that the intrinsic tear performance of Inventive Example 3 was substantially higher than predictable according to the "blend rule" and equivalent to a same-comonomer polymer blend (comparative run 3).

20           FIG. 7 and FIG. 8 show that the intrinsic tear performance of Inventive Example 1 and Inventive Example 2 are substantially comparable to that predictable by the "blend rule".

FIG. 9 shows that at an overall composition density of 0.8992 g/cm<sup>3</sup>, a polymer blend comprised of two interpolymers, each having a different  
25 comonomer (comparative run 4) did not result in a final combination that exhibited good toughness. The intrinsic tear of comparative run 4 was substantially lower than that predicted by the "blend rule" and substantially lower than that of a polymer blend comprised of two interpolymers, each comprising a higher  $\alpha$ -olefin comonomer (comparative run 5).

**We claim:**

1. A diverse interpolymer composition comprising
  - (A) from 5 percent (by weight of the total composition) to 95 percent (by weight of the total composition) of at least one first ethylene interpolymer,  
5 and
  - (B) from 95 percent (by weight of the total composition) to 5 percent (by weight of the total composition) of at least one second ethylene interpolymer;  
10 wherein the first interpolymer is characterized as comprising a different comonomer relative to the second interpolymer;  
wherein one interpolymer is a homogeneously branched ethylene interpolymer and the other interpolymer is a heterogeneously branched ethylene interpolymer or a homogeneously branched ethylene interpolymer,  
and  
15 wherein the composition is characterized as having a density greater than or equal to 0.90 g/cm<sup>3</sup>.
2. The composition according to Claim 1 wherein the first  
interpolymer is a homogeneously branched linear ethylene interpolymer.  
20
3. The composition according to Claim 2 wherein the  
homogeneously branched ethylene interpolymer has a slope of strain  
hardening coefficient greater than or equal to about 1.3.
- 25 4. The composition according to Claim 1 wherein the second  
interpolymer is heterogeneously branched ethylene interpolymer.
5. The composition according to Claim 4 wherein the  
heterogeneously branched ethylene interpolymer is a heterogeneously  
30 branched linear ethylene interpolymer.

6. The composition according to Claim 1 wherein the first interpolymer is a substantially linear ethylene interpolymer characterized as having:

- 5 (a) a melt flow ratio,  $I_{10}/I_2$  5.63,
- (b) a molecular weight distribution,  $M_w/M_n$ , as determined by gel permeation chromatography and defined by the equation:
- $$(M_w/M_n) \leq (I_{10}/I_2) - 4.63,$$
- 10 (c) a gas extrusion rheology such that the critical shear rate at onset of surface melt fracture for the substantially linear ethylene interpolymer is at least 50 percent greater than the critical shear rate at the onset of surface melt fracture for a comparative homogeneously branched linear ethylene interpolymer, wherein the substantially linear ethylene interpolymer and the
- 15 homogeneously branched linear ethylene interpolymer comprise the same comonomer or comonomers, the homogeneously branched linear ethylene interpolymer has an  $I_2$  and  $M_w/M_n$  within ten percent of the substantially linear ethylene interpolymer and wherein the respective critical shear rates of the
- 20 interpolymers are measured at the same melt temperature, under the same nitrogen load using a gas extrusion rheometer, and
- (d) a single differential scanning calorimetry, DSC, melting peak between
- 30° and 150°C.

25

7. The composition according to Claim 1 wherein the first interpolymer is a homogeneously branched linear ethylene/ $\alpha$ -olefin copolymer.

8. The composition according to Claim 1 wherein the first  
30 interpolymer is a substantially linear ethylene/ $\alpha$ -olefin copolymer.



9. The composition according to Claim 1 wherein the second interpolymer is a heterogeneously branched ethylene/ $\alpha$ -olefin copolymer.

5 10. The composition according to Claim 1 wherein the first interpolymer or second interpolymer is a copolymer of ethylene and 1-octene and the other interpolymer is a copolymer of ethylene and 1-pentene.

10 11. The composition according to Claim 1 wherein the first interpolymer or second interpolymer is a copolymer of ethylene and 1-octene and the other interpolymer is a copolymer of ethylene and 1-butene.

15 12. The composition according to Claim 1 wherein the first interpolymer or second interpolymer is a copolymer of ethylene and 1-octene and the other interpolymer is a copolymer of ethylene and 1-hexene.

13. The composition according to Claim 1 wherein the first interpolymer or second interpolymer is a copolymer of ethylene and 1-octene and the other interpolymer is a copolymer of ethylene and 1-heptene.

20

14. The composition according to Claim 1 wherein the composition is further characterized as having an intrinsic tear value greater than or equal to 150 grams, as determined in accordance with ASTM D-1922, Method A.

25 15. The composition according to Claim 1 wherein the composition is further characterized as having an intrinsic tear value greater than or equal to 200 grams, as determined in accordance with ASTM D-1922, Method A.

16. The composition according to Claim 1 wherein the composition is further characterized as having a slope of strain hardening coefficient (SHC) greater than or equal to 0.85.

5 17. The composition according to Claim 1 wherein the composition is further characterized as having a slope of strain hardening coefficient (SHC) greater than or equal to 0.95.

18. A diverse interpolymer composition comprising  
10 (A) from 5 percent (by weight of the total composition) to 95 percent (by weight of the total composition) of at least one first ethylene interpolymer, and

(B) from 95 percent (by weight of the total composition) to 5 percent (by weight of the total composition) of at least one second ethylene  
15 interpolymer;

wherein the first interpolymer is characterized as comprising a different comonomer relative to the second interpolymer and as having a lower density relative to the second interpolymer; the second interpolymer is characterized as having a density less than or equal to 0.95 g/cm<sup>3</sup>;

20 wherein one interpolymer is a homogeneously branched ethylene interpolymer and the other interpolymer is a heterogeneously branched ethylene interpolymer or a homogeneously branched ethylene interpolymer; and

wherein the composition is characterized as having a density in the  
25 range of from greater than or equal to 0.90 g/cc to about 0.935 g/cc.

19. A film comprising the composition of Claim 1.

20. A fiber comprising the composition of Claim 1.

30

21. A molding comprising the composition of Claim 1.
22. A coating comprising the composition of Claim 1.
- 5 23. A profile comprising the composition of Claim 1.
24. A pouch comprising the composition of Claim 1.
25. A sealant film layer comprising the composition of Claim 1.
- 10 26. A carpet backing comprising the composition of Claim 1.
27. A liner comprising the composition of Claim 1.
- 15 28. A shrink film comprising the composition of Claim 1.
29. A stretch film comprising the composition of Claim 1.
30. An extrusion coating comprising the composition of Claim 1.
- 20 31. A laminating film comprising the composition of Claim 1.
32. A rotomolding comprising the composition of Claim 1.
- 25 33. A sack comprising the composition of Claim 1.
34. The sack according to Claim 33 wherein the sack is a grocery sack.
- 30 35. A bag comprising the composition of Claim 1.

36. The bag according to Claim 35 wherein the bag is a merchandise bag.

5 37. The pouch according to Claim 24 wherein the pouch is fabricated using form-fill-seal (FFS) equipment.

38. The pouch according to Claim 37 wherein the equipment is a vertical form-fill-seal unit.

FIG. 1A

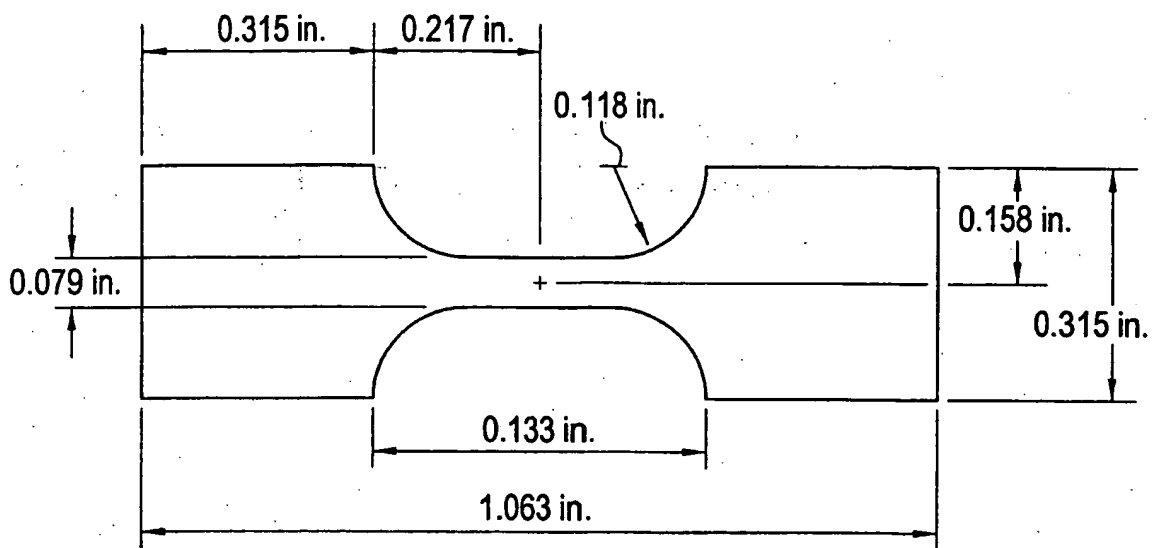


FIG. 1B

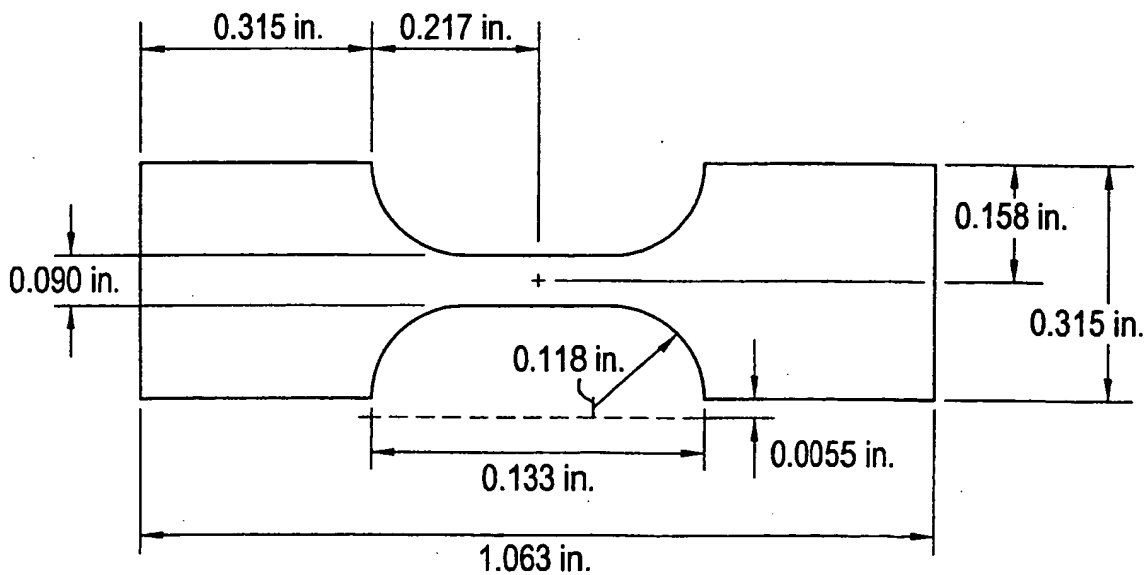
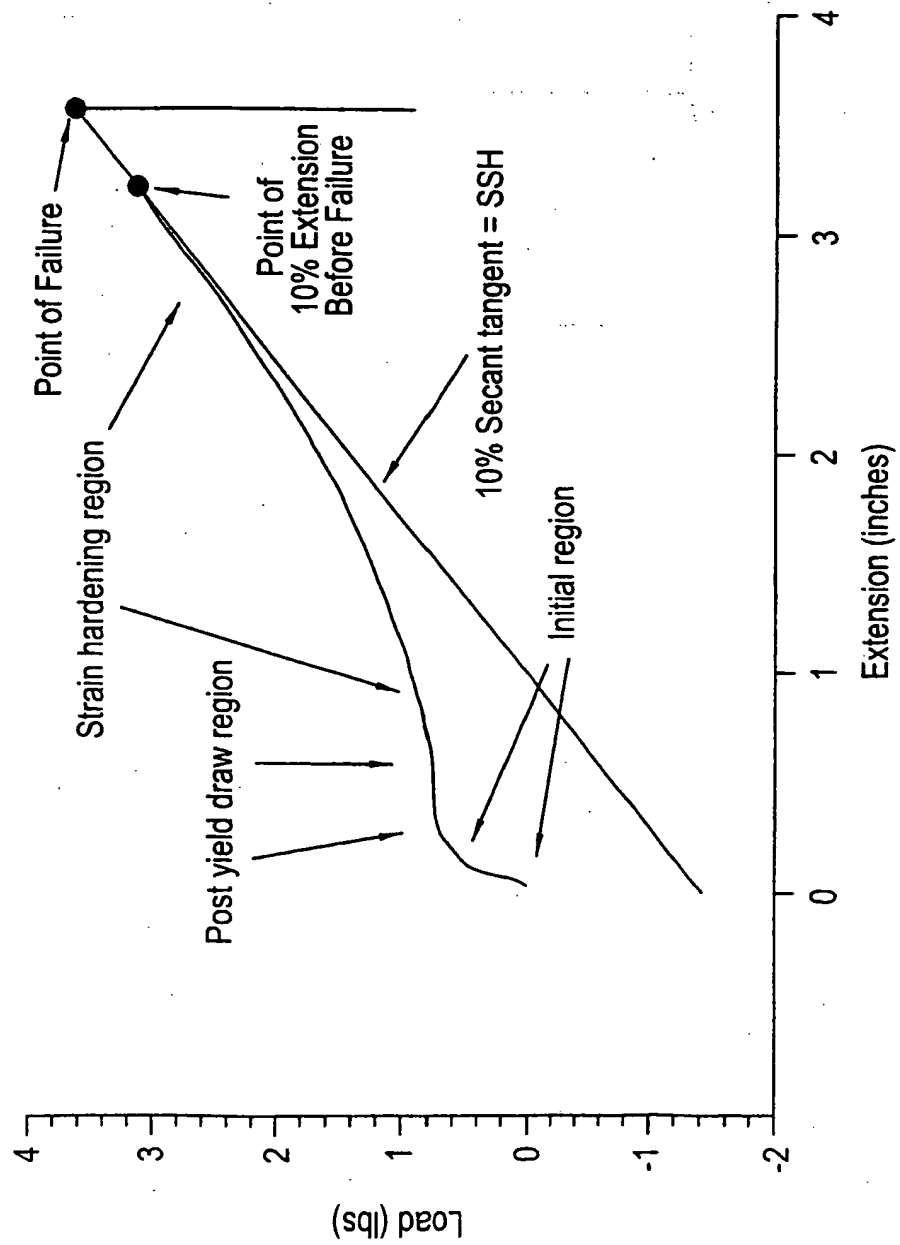


FIG. 2



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FIG. 3

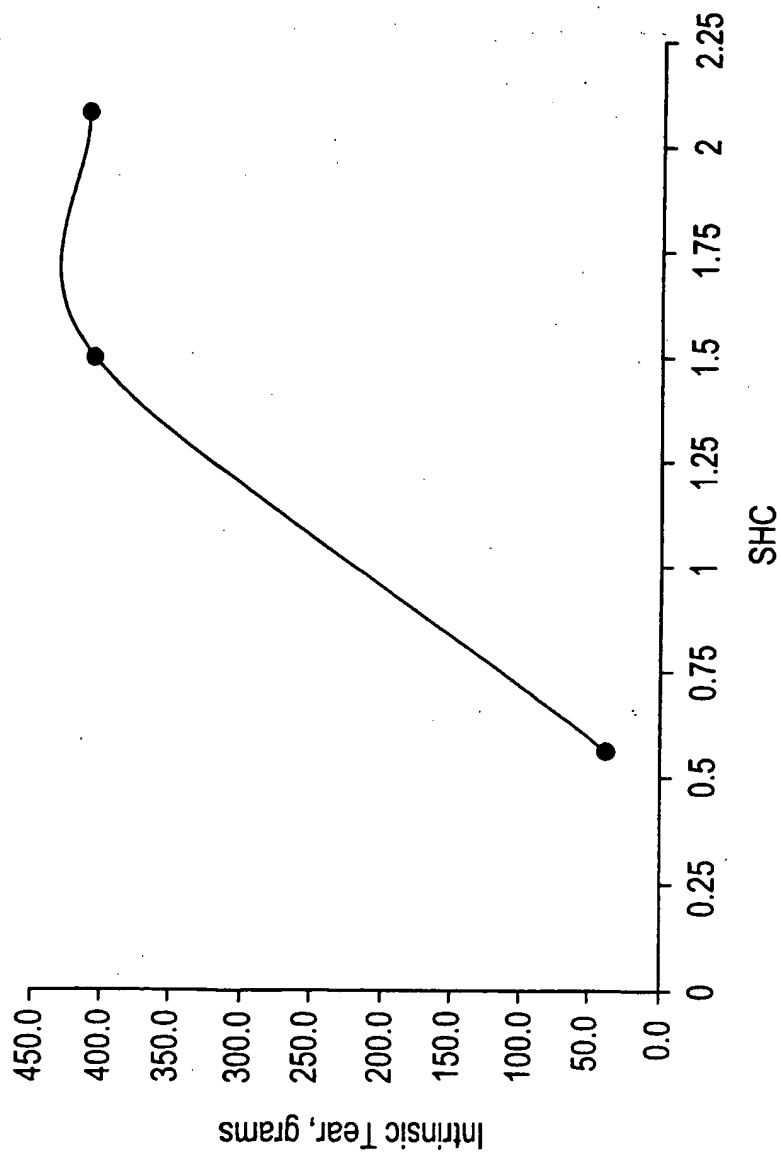
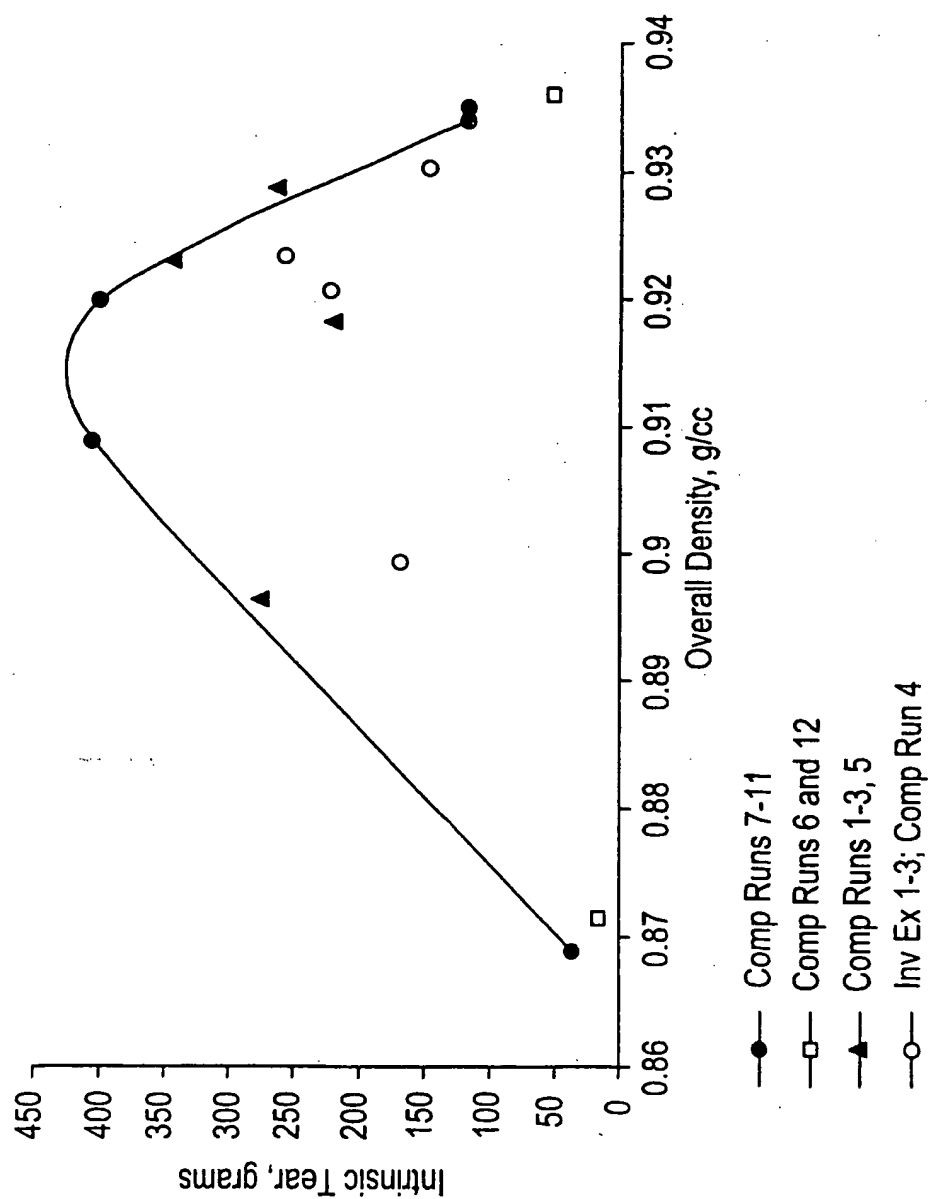


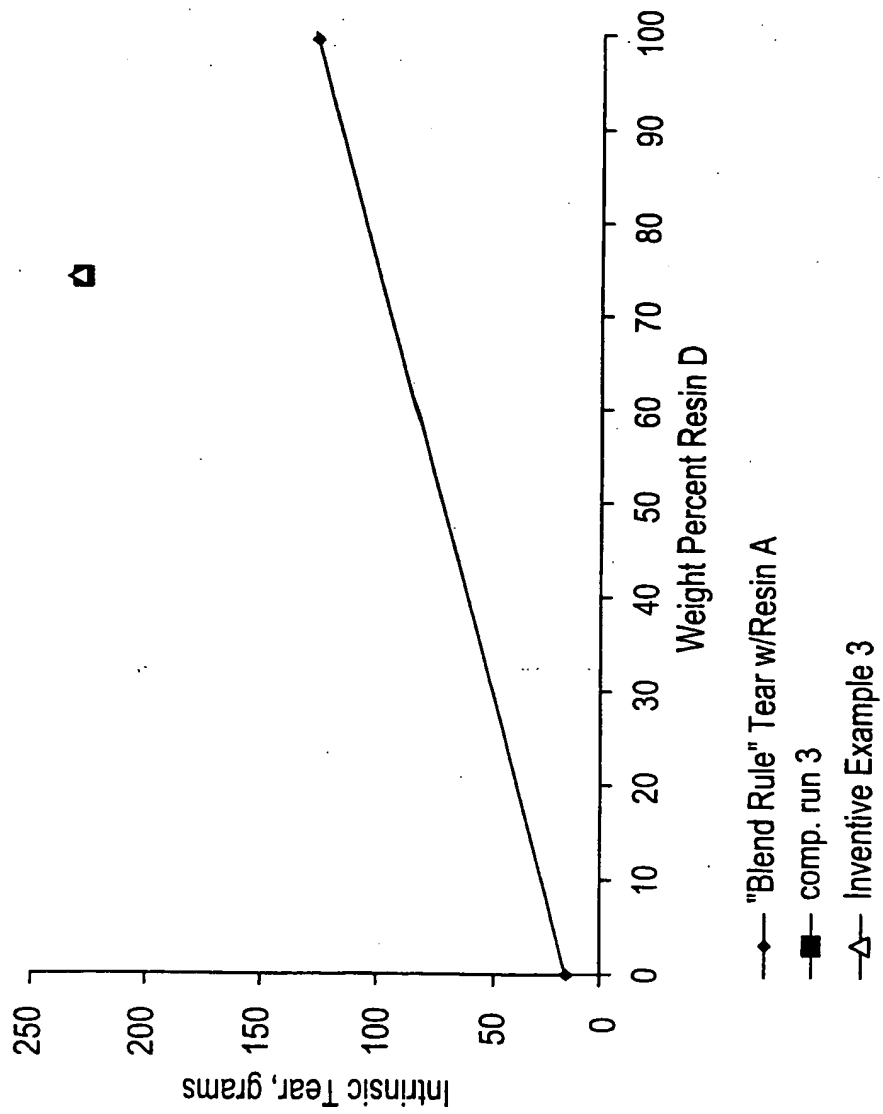
FIG. 4





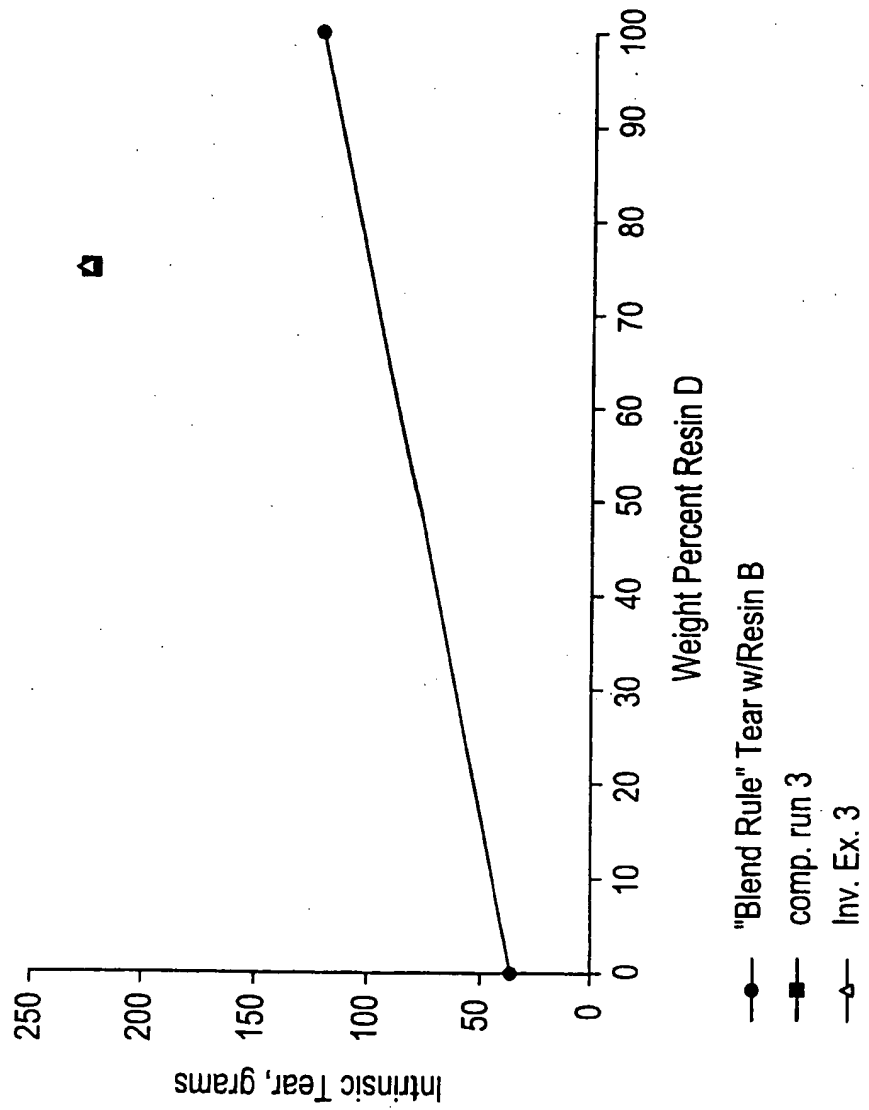
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FIG. 5



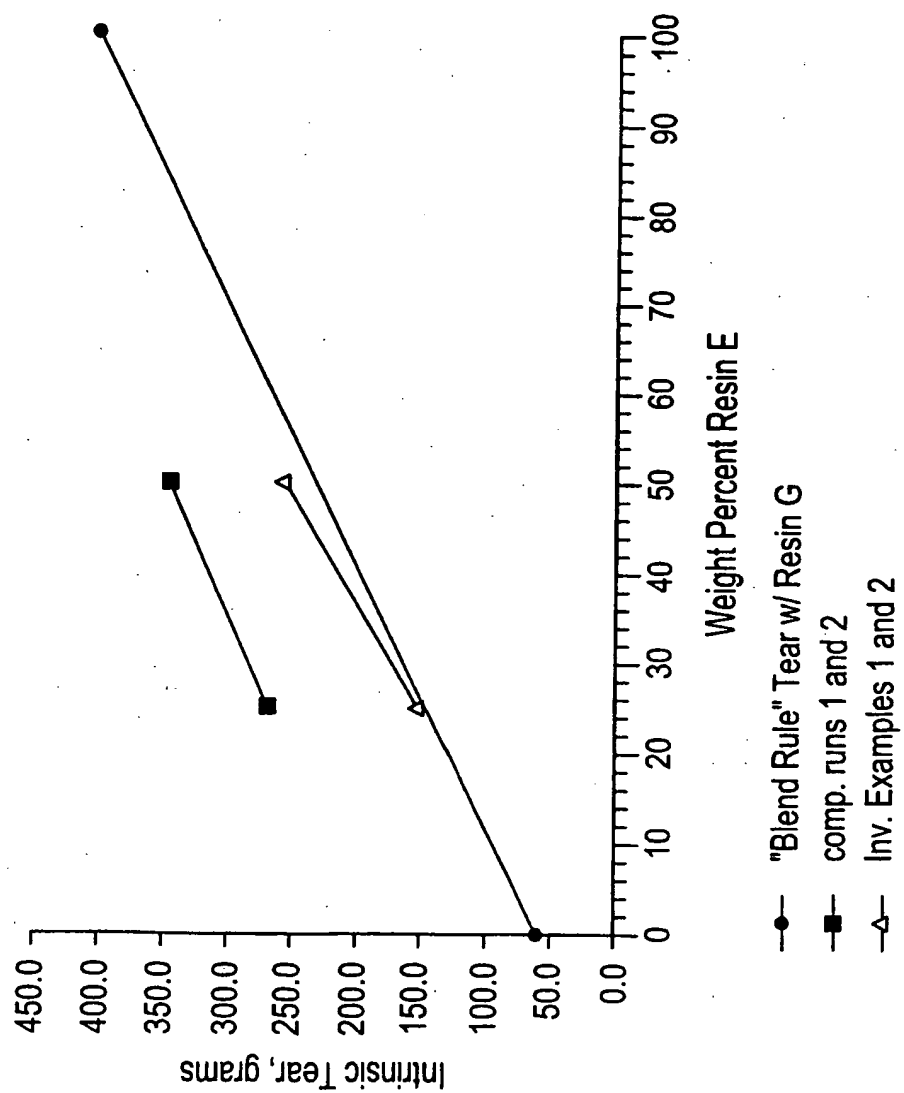
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FIG. 6



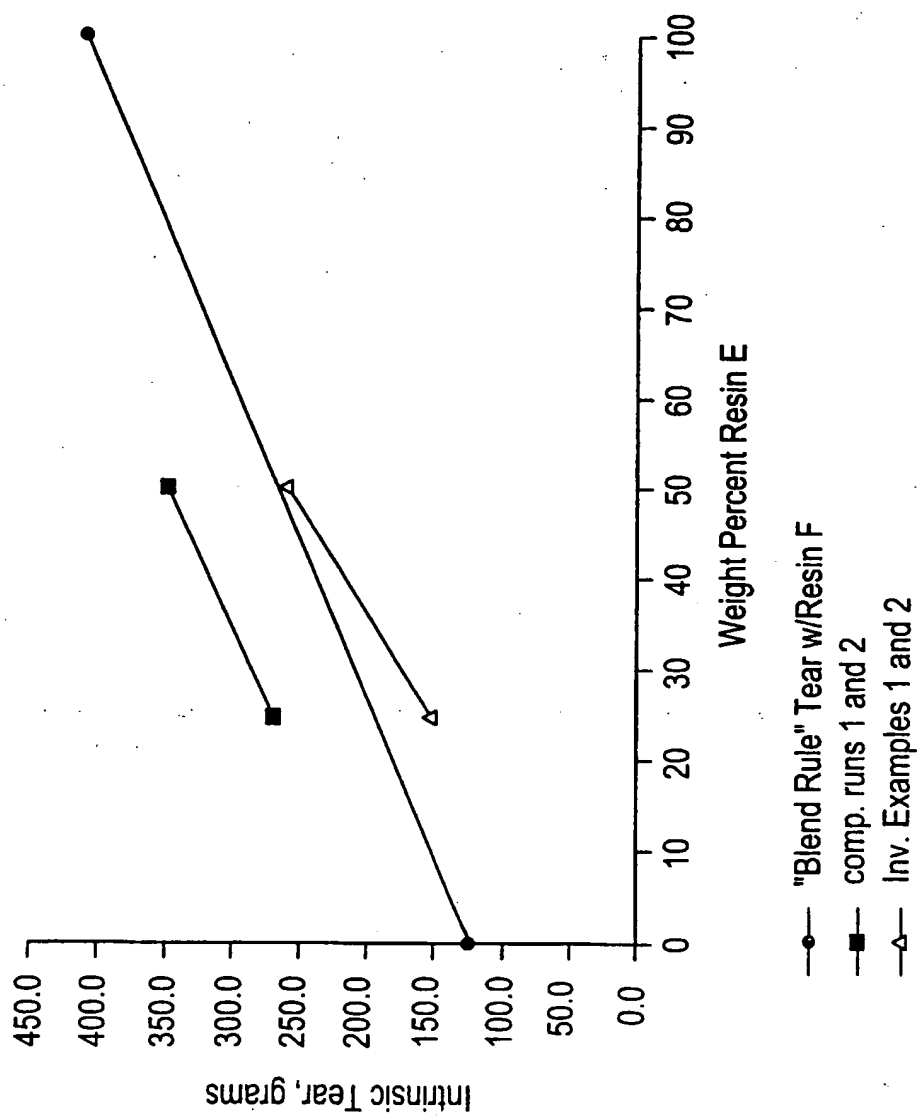
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FIG. 7



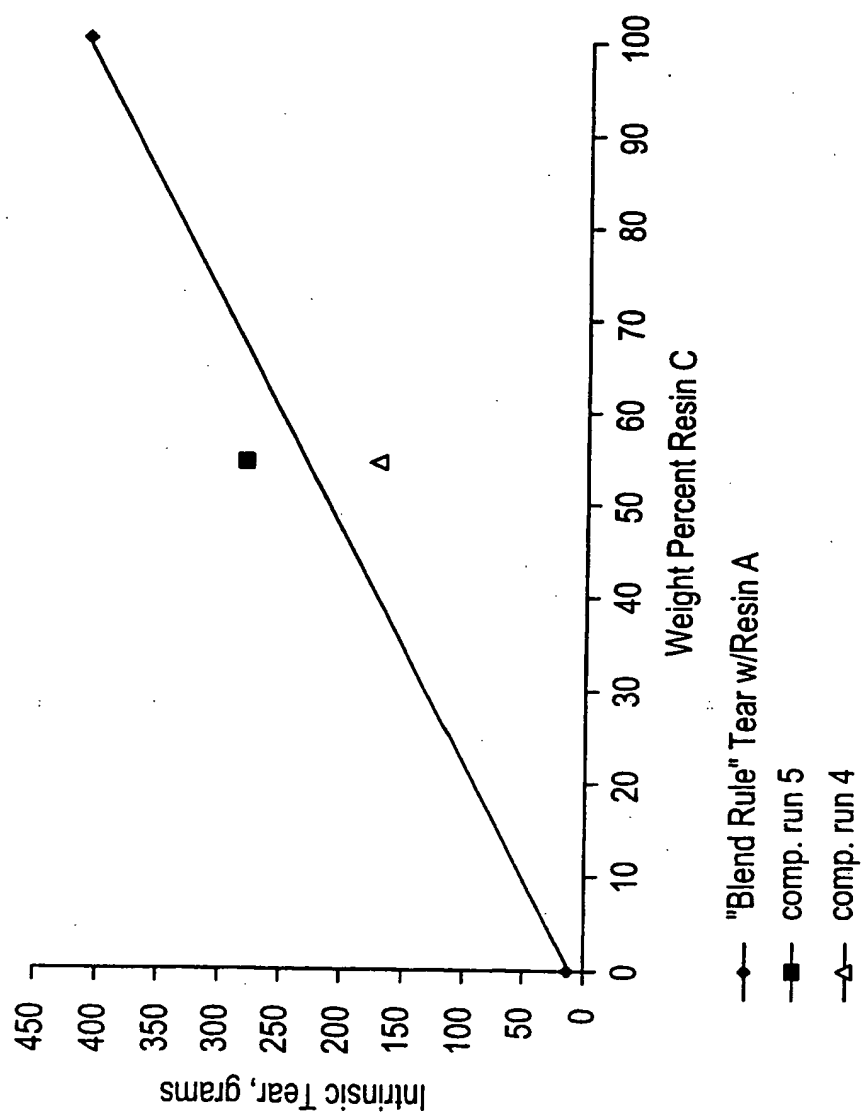
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FIG. 8



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FIG. 9



# INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 00/30093

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C08L23/04 C08J5/18 C09D123/04

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C08L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

WPI Data, PAJ, EPO-Internal

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	<p>WO 94 25523 A (DOW CHEMICAL CO)  10 November 1994 (1994-11-10)  cited in the application  abstract; claims 1-16  page 5, line 14-26  page 8, line 3-5,8-10  page 9, line 11  page 12, line 6  page 13, line 25  page 14, line 5,14,32  page 15, line 25</p> <p style="text-align: center;">--- -/-</p>	1-5,7-9, 11-38



Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

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Date of the actual completion of the international search

1 February 2001

Date of mailing of the international search report

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# INTERNATIONAL SEARCH REPORT

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## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

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A	<p>page 4, line 26 page 7, line 17</p>	<p>2-38</p>
A	<p>WO 90 03414 A (EXXON CHEMICAL PATENTS INC) 5 April 1990 (1990-04-05) abstract; claims 1-26 page 6, line 14-30 page 7, line 1-5,28-31</p>	<p>1-38</p>

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